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Exploiting the Negative Polarization Properties of Indium Gallium Nitride (InGaN)/Gallium Nitride (GaN) Heterostructures to Achieve Frequency Doubled Blue-green Lasers with Deep UV (<250 nm) Emission (Year 2)

by Dr. Meredith Reed and Dr. Eric D. Readinger

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14. ABSTRACT

There is an Army need to develop deep ultraviolet (UV) semiconductor lasers that are compact with a low power budget for use in real-time reagentless biodetection and identification systems as well as water monitoring. Our approach is to develop a visible indium gallium nitride (InGaN)-based laser that exploits the negative polarization charge at the heterointerface, which can then be frequency doubled into the deep UV. In the second year of this Director's Research Initiative (DRI), we focused on the growth and development of periodically poled AlGaN to be used for frequency doubling the visible laser light into the deep UV (DUV). To accomplish this, we have demonstrated the successful growth of inverted, N-polar AlN, which was achieved by Mg overdoping using a larger inversion layer thickness than for N-polar GaN. We have demonstrated a sub-micron periodic poled AlN by e-beam lithography, ICP etching, and MBE regrowth.

15. SUBJECT TERMS

III-Nitrides, Semiconductor, GaN, Polarization, Piezoelectric, InGaN, Water Purification and Monitoring, UV

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1. Statement of the Army Problem

There is an Army need to develop deep ultraviolet (UV) semiconductor lasers that are compact with a low power budget, for use in real-time reagentless biodetection and identification systems as well as water monitoring systems which require a laser in the fluorescence free regime (<250 nm) for UV resonance Raman spectroscopy. The Edgewood Chemical and Biological Center (ECBC) has stated a need for deep UV lasers for bio-threat detection systems, while the Tank and Automotive Research, Development and Engineering Center (TARDEC) has expressed a need for these detection systems as well as blue-green lasers for water monitoring and purification systems. At present, deep UV semiconductor lasers have not been realized, with *p*-type doping and suitable substrate/templates as two areas of research that still require maturation. In addition, blue-green lasers have not been developed due to the detrimental effects of spontaneous and piezoelectric polarization in nitride semiconductors, which leads to efficiency droop at high injection current due to carrier leakage and enhanced Auger recombination in the quantum wells (QWs). The development of high power green lasers will also permit advancement in high power, high efficiency green light emitting diodes (LEDs) necessary for solid-state lighting solutions for landing beacons and white light illuminators at check points. Therefore, we propose to address both of these needs by utilizing the benefits of negative polarization in indium gallium nitride (InGaN)/gallium nitride (GaN) heterostructures.

2. Objectives

The objective of this research is to explore, develop and demonstrate the feasibility of exploiting the negative polarization properties of InGaN/GaN heterostructures to achieve frequency doubled blue-green lasers with deep UV (<250 nm) emission.

3. Approach to Achieving the Objectives with Milestones

III-nitride semiconductor lasers in the UV have not been realized due to difficulties associated with the inability to sufficiently *p*-type dope $Al_xGa_{1-x}N$ layers with high aluminum (Al) compositions (x>0.5) and the lack of substrates for growth of $Al_xGa_{1-x}N$ active layers. Therefore, the development of an InGaN blue-green laser that can be frequency doubled allows for deep UV lasers that meet current Army needs. However, the path to blue-green high power LEDs and lasers is also difficult given that these devices suffer from efficiency droop at high injection current, which results from polarization charges at the hetero-interface. Typical III-nitride

heterostructure devices are grown along the [0001] direction (Ga-polar) with a p-GaN cap ("*p*-up"), which result in strong built-in electric fields associated with large spontaneous and piezoelectric positive polarization charges at the hetero-interfaces. The positive polarization charge combined with a small conduction band offset in the InGaN/GaN junction leads to a reduced barrier for electron injection, allowing for carrier leakage into the *p*-GaN, while increasing the barrier height for hole injection. To reduce the electron overshoot, an AlGaN electron-blocking layer with a larger bandgap must be used, which increases the polarization field in the quantum well and effectively blocks the injection of holes (*1*). One approach to circumvent these polarization charges and the need for an AlGaN blocking layer is to grow non-polar GaN devices on non-polar substrates (*2, 3*); however, non-polar substrates are very expensive and not well lattice matched, and there are difficulties incorporating appropriate In content for green emission. However, the positive polarization charges at the hetero-interface of [0001] (In)GaN materials can be ameliorated by reversing the growth sequence of the layers ("*p*-down") or by inverting the polarity (N-polar). This results in a negative polarization charge at the hetero-interface increasing the electron barrier, lowering the hole barrier, and creating a two dimensional hole gas (2DHG) within the *n*-InGaN. Another approach to create a negative polarization charge at the hetero-interface is the use of semi-polar InGaN/GaN devices (*4*). With the advent of thick, *p*-type as-grown GaN templates on sapphire ("*p*-down"), N-polar GaN substrates, and semi-polar (11–22) GaN templates it is now possible to pursue laser heterostructures that utilize the benefits of negative polarization charges.

4. Results

In the first year, we grew and characterized bulk InGaN epilayers with a compositional range up to 33% In for application as *n*-InGaN active regions on various substrates. We fabricated single-heterostructure (SH) n-$In_xGa_{1-x}N$/p-GaN ("*p*-down") LED structures grown on sapphire substrates with In compositions varied from $0.07<x<0.33$. Standard device processing techniques created a mesa in the 0.1–0.2 µm thick InGaN active layer, which defined the device active area. The devices were diced, mounted on TO headers and tested for output power and wavelength as a function of current density. Figure 1(a) shows the typical emission spectra and output power at 10% duty cycle current densities for a Ga-polar *p*-down SH with 22% In composition, having a peak intensity at 485 nm (blue-green).

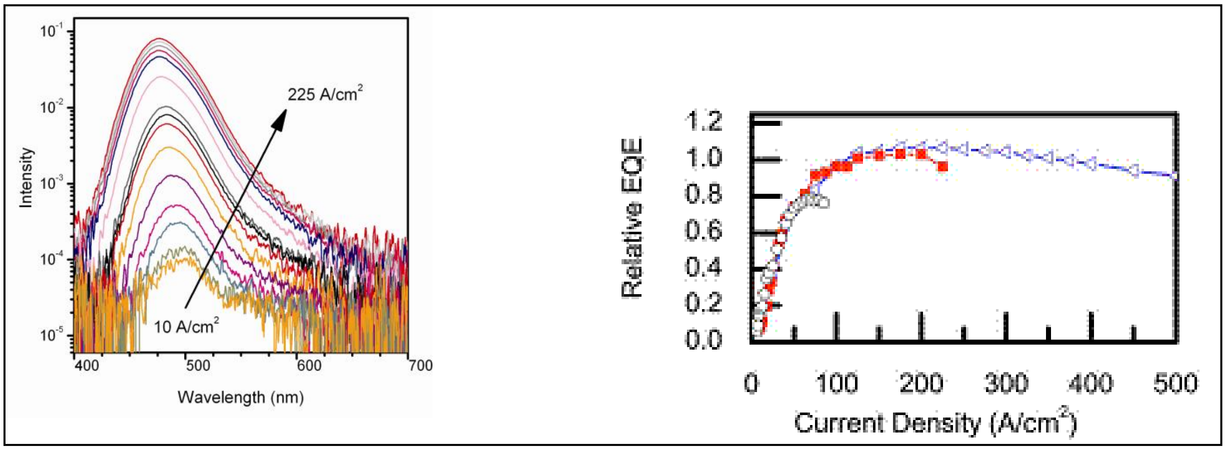

Figure 1. (a)The typical emission spectra at 10% duty cycle with respect to current density for an *n*-$In_{0.22}GaN$/*p*-GaN *p*-down, Ga-polar device, and (b) the relative external quantum efficiency (EQE) as a function of current density for cw (circles), 1% (triangles) and 10% (squares) duty cycle.

The EQE of this device as a function of duty cycle is shown in figure 1(b). Under 1% duty cycle the current injection peaks above 100 A/cm^2, well beyond the current density 10–25 A/cm^2 at which conventional *p*-up, Ga-polar InGaN/GaN multi-quantum well LEDs exhibit significant efficiency droop (*1, 5*). The achievement of a blue-green LED with reduced efficiency droop at high current densities has been a significant achievement in the area of visible emitters, and is a necessary step for achieving a blue-green laser that makes use of the benefits of the negative polarization charge.

During the second year of this Director's Research Initiative (DRI), the following technical areas were explored to accomplish the proposed objectives. Work continued on making device structures for solid-state lasers using a variety of different approaches. The work on this DRI for the second year focused on the growth and development of periodically poled AlGaN to be used for frequency doubling the visible laser light into the deep UV (DUV).

4.1 Development of Materials for Frequency Doubling Green-Blue Laser

Another aspect of the project that was explored was our approach to frequency doubling the laser to achieve a DUV laser. The idea is to employ a second-harmonic generation (SHG) using the visible laser source and a periodically poled waveguide structure to facilitate frequency doubling into the UV and DUV. The use of first-order quasi-phase matching for SHG from visible to DUV by periodic poling in nonlinear materials is advantageous as it allows increased interaction length and conversion efficiency in nontraditional, nonlinear materials transparent to the DUV. Unlike materials with longer interaction lengths, which can be used outside the laser, materials with shorter interaction length, such as beta-barium borate (BBO), must be contained inside the lasing cavity. Periodically poled GaN has been used for SHG in the infrared spectrum; however, successful operation into the DUV requires a wider bandgap (*6*). Aluminum nitride (AlN) is of particular interest for SHG of deeper UV wavelengths by quasi-phase matching for a few

reasons. First, AlN can be practically periodically poled by e-beam lithography, inductively coupled plasma (ICP) etching and molecular beam epitaxy (MBE) regrowth. Second, AlN is more transparent than typical nonlinear materials such as potassium di-hydrogen phosphate (KDP) and lithium triborate (LBO) and can be readily employed for SHG and deeper UV.

Frequency doubling can be achieved by using a pump wave from a low frequency (laser) source generates a new, nonlinear polarization wave with twice the frequency in a nonlinear crystal—i.e., second-harmonic generation—by way of the second-order nonlinear $X^{(2)}$ susceptibility. The second-harmonic wave is aligned in the direction of the pump wave by quasi-phase matching (QPM) from the nonlinear crystal—here, periodically poled AlN (figure 2).

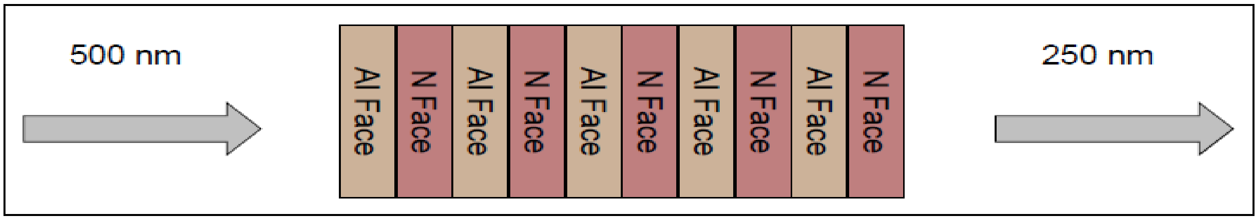

Figure 2. Frequency doubling concept using periodically poled AlN (nonlinear crystal).

With QPM, a material with spatially modulated nonlinear properties is used rather than a single, homogeneous nonlinear crystal. In these special materials, a small phase mismatch is allowed for a certain propagation distance, but reversed at positions where interaction would be destructive. Using QPM, rapid growth of the second harmonic is obtained by changing the polarity (P_S) throughout the nonlinear crystal (see curve B of figure 3). Although QPM does not increase the amplitude of the SHG wave as quickly as for perfect phase matching (curve A), it can be used with a much wider range of nonlinear materials and at lower (i.e., room) temperatures and without worry of spatial walk-off.

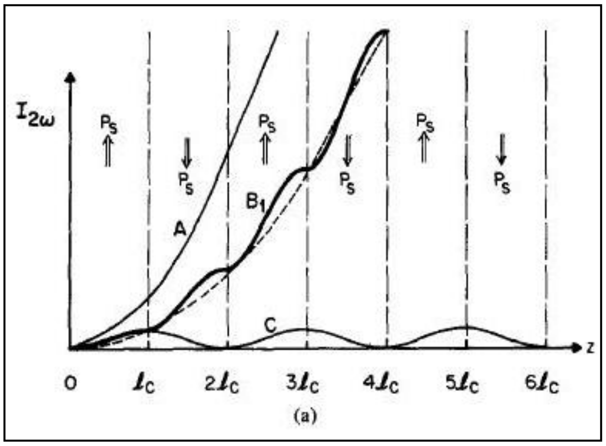

Figure 3. Effect of phase matching on the growth of second-harmonic intensity with distance in a nonlinear crystal: A: perfect phase matching in a uniformly poled crystal, C: nonphase-matched interaction, and B_1: first-order QPM by flipping the sign of the spontaneous polarization every coherence length of the interaction of curve C (*7*).

Efficient frequency doubling by QPM requires a constant phase relationship over the course of the nonlinear crystal, in this case AlN. For SHG by first-order QPM, the grating periodicity Λ is given as (*6*)

$$\Lambda = \frac{\lambda_\omega}{2(n_{2\omega} - n_\omega)}, \tag{1}$$

where λ_ω is the fundamental wavelength at ω, and n_ω and $n_{2\omega}$ are the refractive indices at the fundamental and second-harmonic wavelengths, respectively. The required period for a given second harmonic is dependent on material parameters including refractive index and anisotropic dielectric function, as shown in figure 4; λ's of interest (particularly, below 260 nm) require a period of below 2000 nm (half period 1000 nm)

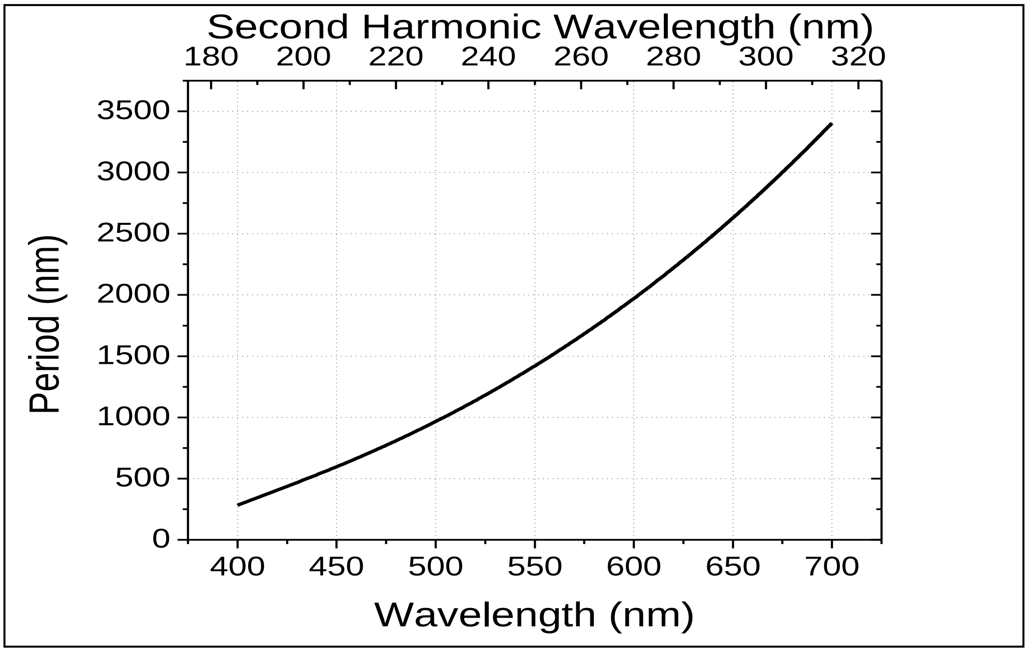

Figure 4. Shows the required period for a given second harmonic for a given λ.

The experimental process (shown in figure 5) is as follows we grow an inverted, N-polar AlN is grown by MBE via magnesium (Mg) overdoping on an Al-face AlN template. E-beam lithography is used to pattern a dense, periodic structure of symmetric, equal width trench and ridge. Half periods between1 μm and 150 nm are explored. The structure is then etched down to the Al-face AlN layer and regrown resulting in a periodically poled waveguide.

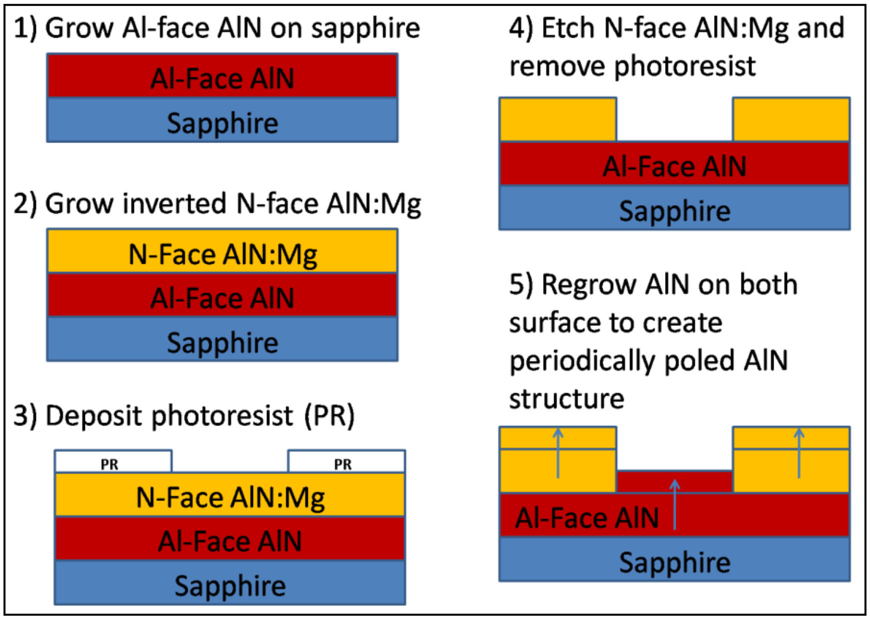

Figure 5. Experimental process for creating a periodically poled AlN structure for frequency doubling.

In order to achieve the periodically poled AlN, MBE was used as the growth technique. Polar inversion of GaN is achieved by Mg overdoping of III-nitrides during MBE growth, which is well documented (*8, 9*). Submicron period poled-AlN structures are fabricated by initially growing an Al polar AlN film by MBE at a substrate temperature of 900 °C and then inverting the polarity of the film by Mg overdoping. Polarity inversion was confirmed in-situ by observing a 3× surface reconstruction by reflection high-energy electron diffraction (RHEED) after deposition of the Mg overdoped layer, as shown in figure 6.

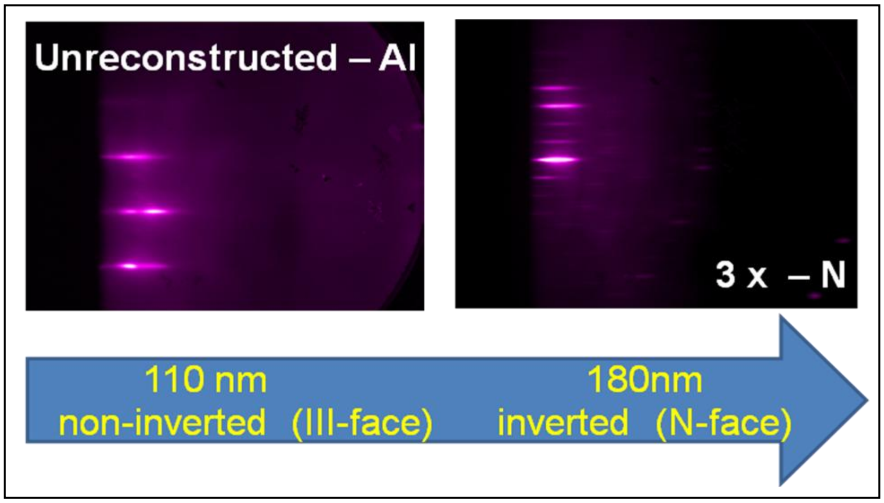

Figure 6. RHEED pattern showing inversion from Ga-face to N-face during MBE growth.

Next, e-beam lithography was used to define a nickel (Ni) hard etch mask consisting of stripes having spacings between 0.15 and 1 μm. Side-wall angle of the periodic mesa structures, surface roughness of the etched III-face, and overall etch rate were characterized as a function of ICP etch parameters, including pressure, radio frequency (RF) bias, ICP power, and gas composition. Crystallographic orientation of the growth surface is known to have an effect on surface morphology and faceting. As a result, stripes were aligned along either the m- (1010) or a- (1120) directions. Surface morphology and etch depth were investigated by scanning electron microscopy (SEM) and atomic force microscopy (AFM). Average initial root-mean squared (RMS) roughness of the AlN film was 2.25 nm. Finally, the patterned film was regrown by plasma-assisted molecular beam epitaxy (PAMBE) at different III-V flux ratios to produce a periodically poled AlN structure, as shown in figure 7.

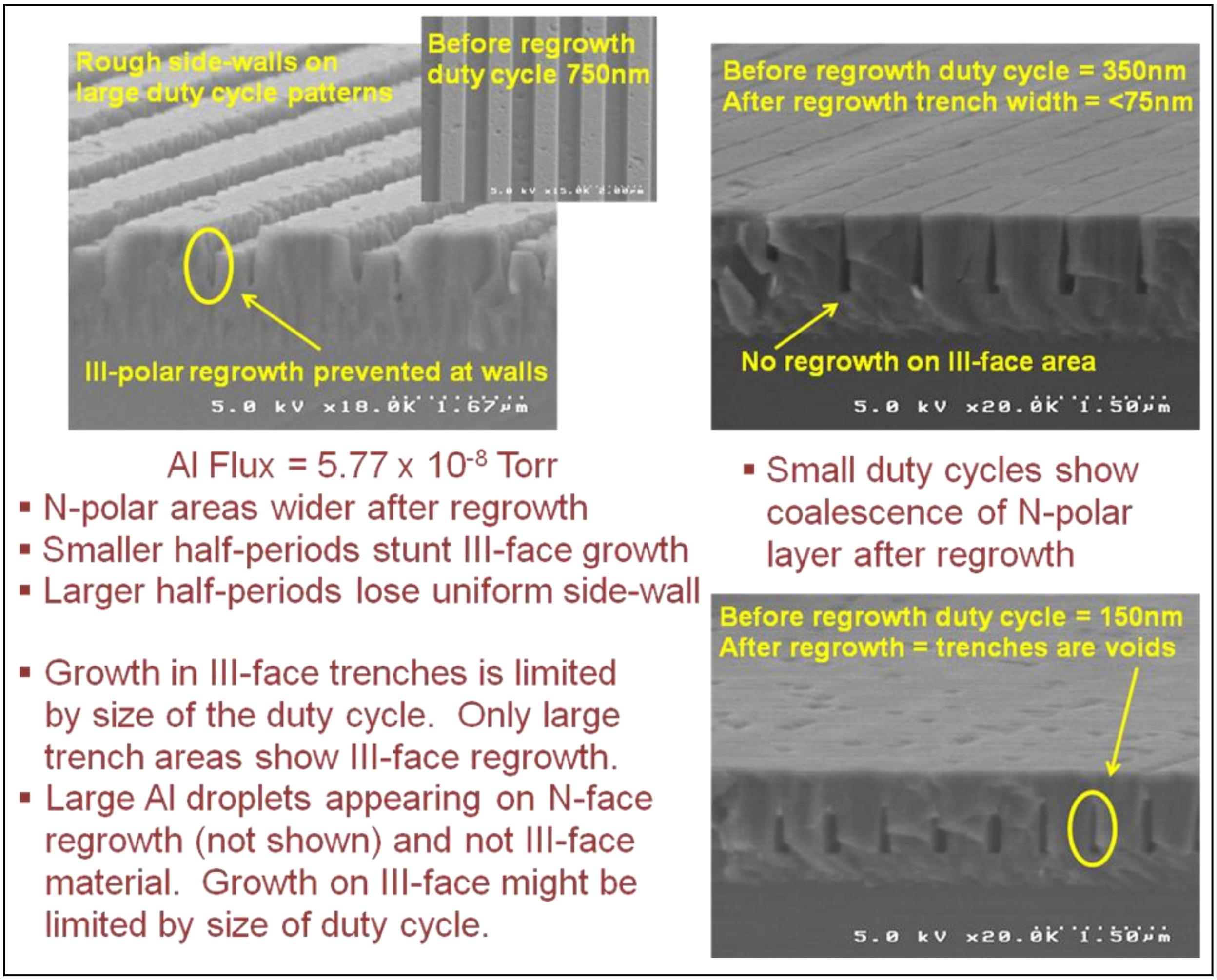

Figure 7. SEM images showing before and after regrowth in creating a periodically poled AlN structure for frequency doubling.

5. Conclusions

We have demonstrated the successful growth of inverted, N-polar AlN, which was achieved by Mg overdoping using a larger inversion layer thickness than for N-polar GaN. We have demonstrated a sub-micron periodic poled AlN by e-beam lithography, ICP etching, and MBE regrowth. Our path forward is to optimize the waveguide parameters so that they are large enough to collect the laser beam, but small enough for lateral confinement to the light path. We will continue working on the laser structures for frequency doubling as part of the upcoming Defense Advanced Research Projects Agency (DARPA) program for work in the DUV.

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7. Transitions

The LED portion of this work is being funded through a Department of Energy (DOE) program to address the “green” gap problem, $1.8M over 3 years. The development of a frequency doubled laser into the DUV will have immediate applications in communications, water purification and monitoring, and bio-threat sensing.

As part of the FY10 DRI, we delivered two conference presentations and papers at the DOE Solid-state Lighting Workshop and the International Workshop on Nitrides. We also produced one journal publication in *Physica Status Solidi* and filed one patent.

List of Symbols, Abbreviations, and Acronyms

AFM	atomic force microscopy
Al	aluminum
AlN	aluminum nitride
BBO	beta-barium borate
DARPA	Defense Advanced Research Projects Agency
DOE	Department of Energy
DRI	Director's Research Initiative
DUV	deep UV
ECBC	Edgewood Chemical and Biological Center
EQE	external quantum efficiency
GaN	gallium nitride
ICP	inductively coupled plasma
InGaN	indium gallium nitride
KDP	potassium di-hydrogen phosphate
LBO	lithium triborate
LEDs	light emitting diodes
MBE	molecular beam epitaxy
Mg	magnesium
Ni	nickel
PAMBE	plasma-assisted molecular beam epitaxy
QPM	quasi-phase matching
QW	quantum well
RF	radio frequency
RHEED	reflection high-energy electron diffraction

RMS	root-mean squared
SEM	scanning electron microscopy
SH	single heterostructure
SHG	second-harmonic generation
TARDEC	Tank and Automotive Research, Development and Engineering Center
UV	ultraviolet

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